Space-Time Approach to Scattering from Many Body Systems

R. Gähler*, J. Felber*, F. Mezei# and R. Golub#
#Hahn Meitner Institut, Glienickerstr. 100, D-14109 Berlin
*TU-München, Fakultät für Physik, D-85747 Garching

February 2, 2008

Abstract

We present scattering from many body systems in a new light. In place of the usual van Hove treatment, (applicable to a wide range of scattering processes using both photons and massive particles) based on plane waves, we calculate the scattering amplitude as a space-time integral over the scattering sample for an incident wave characterized by its correlation function which results from the shaping of the wave field by the apparatus. Instrument resolution effects - seen as due to the loss of correlation caused by the path differences in the different arms of the instrument are automatically included and analytic forms of the resolution function for different instruments are obtained. Each element of the apparatus is associated with a correlation length (or time). These correlation lengths, determined by the dimensions of the apparatus are generally much smaller than these dimensions and larger than the wavelength. As is well known, these are the conditions for the validity of geometrical optics so that the conventional treatment, where the scattering is calculated by the van Hove plane wave approach and the trajectories through the instrument are treated classically, is usually valid. In the present approach analytic expressions for the correlation functions are obtained. The intersection of the moving correlation volumes (those regions where the correlation functions are significant) associated with the different elements of the apparatus determines the maximum correlation lengths (times) that can be observed in a sample, and hence, the momentum (energy) resolution of the measurement. This geometrical picture of moving correlation volumes derived by our technique shows how the interaction of the scatterer with the wave field shaped by the apparatus proceeds in space and time. Matching of the correlation volumes so as to maximize the intersection region yields a transparent, graphical method of instrument design. PACS: 03.65.Nk, 3.80 +r, 03.75, 61.12.B

1 Introduction

The usual treatment of scattering from many particle systems assumes the initial beam to be represented by a plane wave and shows that the scattering cross section is proportional to the Fourier transform of the correlation function of the density fluctuations. For inelastic (energy changing) scattering this is the van Hove time dependent correlation function, in the case of elastic scattering it is the static pair distribution function [1]. The usual treatment then integrates the cross section for monochromatic plane waves over the spread of momentum in the incident and scattered beams.

In this work we show that another approach is both possible and interesting. As we do the entire calculation in real space-time our approach sheds new light on the emergence of the Fourier transform in the scattering cross section. A crucial feature of our approach is that the influence of beam preparation on the resolution of the measurement is expressed by the correlation volume of the incident beam, which in turn, is determined by the optical (in the sense of normal [2] and time dependent [3] optics) properties of the beam preparation elements, *e.g.* slits and choppers.

We will see that the lateral width of the beam correlation volume determines the possible spatial extent of the correlations which can be observed in a given experiment, and is inversely proportional to the q resolution, as in the conventional treatment. The longitudinal extent of the beam correlation volume or the correlation time of the incident beam likewise sets the limit to the system correlation times which can be observed. The correlation volume V_c is the conjugate volume to the momentum space volume V_p and $V_p V_c \approx \hbar^3$.

In a previous work [4] we have discussed the neutron spin echo spectrometer [5] from a similar point of view. We have shown that in that case the equivalent correlation volume is split longitudinally by the 'spin echo time', τ_{NSE}, which can be much longer than the correlation time of the incident beam. This splitting of the correlation volume results in the fact that the spin echo spectrometer measures the time dependence of the scattering system correlation function directly. When the incoming beam correlation volume is continuous (not split), then the scattered wave is an integral over all space-time separations falling within the correlation volume of the incident beam and the result is the Fourier transform of the space-time correlation function of the scattering system as will be seen below.

1.1 Correlation functions and scattering

The concept of correlation (coherence) volume has been applied to visible light optics for more than 50 years including such problems as the effect of the illumination on the resolution of a microscope . The correlation properties of a beam as a result of the optical properties of its preparing devices are known as the van Cittert-Zernike theorem [2], [6] and [7] and we will see how this can be applied to both time - dependent and -independent scattering.

A frequently used technique for deconvoluting the instrument resolution from the measured data is to use the Fourier transform of the resolution function [8]. In the present work we show the physical significance of that Fourier transform and show how it is determined by the optical properties of the beam. It has been shown in [8] that this Fourier transform may contain more information than *e.g.* the width of the resolution function. In fact it is the correlation function at long distances and times that is often of the greatest importance. An additional feature of our treatment is that the instrumental resolution is seen as an integral part of the experiment not something that is put in *ad hoc* after the calculation.

It is well known that scattering experiments depend only on the spectrum $\left|A\left(\overrightarrow{k}\right)\right|^2$ of the incident beam and hence can be considered as the incoherent superposition of the results of experiments done with plane waves distributed according to this spectrum. This is easily seen in the context of the present work where we show that the scattering cross section depends on the auto-correlation function of the incident beam. The Fourier transform of this auto-correlation is $\left|A\left(\overrightarrow{k}\right)\right|^2$. In addition it is usually assumed that sources produce 'chaotic' beams, *i.e.* beams with $\left\langle A\left(\overrightarrow{k_1}\right)A^*\left(\overrightarrow{k_2}\right)\right\rangle = C\delta\left(\overrightarrow{k_1}-\overrightarrow{k_2}\right)$, [9], so that in any case it is only $\left|A\left(\overrightarrow{k}\right)\right|^2$ which is significant. This is the case for 'stationary' beams where $\left\langle|\psi(x)|^2\right\rangle$ is independent of x.

These points, and the possibilities of measuring $\left\langle A\left(\overrightarrow{k_1}\right)A^*\left(\overrightarrow{k_2}\right)\right\rangle$ have been discussed in [10] and the references therein, and have been emphasized by Mezei [11]. For these reasons we feel that it is more convenient to talk about correlation volumes rather than coherence volumes as has been the practice in optics [2] since it seems reasonable to reserve the term coherence for those cases where $\left\langle A\left(\overrightarrow{k_1}\right)A^*\left(\overrightarrow{k_2}\right)\right\rangle$ plays an important role.

The correlation length δ_c discussed here and in section 3 has real physical meaning (as of course does its equivalent the spectral width Δk). This can be seen by considering that in order to observe the famous two slit interference pattern (Young) it is necessary to illuminate the two slits with correlated radiation. This is normally produced by having the two slits located in the main peak of the diffraction pattern produced by a third slit up-stream from the pair producing the interference pattern. The van Cittert- Zernike theorem is the rigorous expression of this point. The two point correlation function can be measured with such a double slit experiment by measuring the modulation (visibility) of the interference pattern as a function of the slit separation. Of course the physical situation can be described according to the spectral viewpoint, where the incident beam is considered as an incoherent superposition of different $\overrightarrow{k}$ vectors, by considering that for increasing slit separations the interference patterns for the different incident $\overrightarrow{k}$ vectors begin to cancel out.

Recent experiments involving Mössbauer scattering of synchrotron radiation have been described in terms of the beam correlation volume [12]. These experiments can also be interpreted in terms of $\left|A\left(\overrightarrow{k}\right)\right|^2$. However the authors have shown that the concept of a coherence (correlation) length is very useful for understanding this exciting experiment.

In the following we shall see that these ideas have a rigorous basis. We have presented the main features of these ideas with some physical examples in [13].

1.2 Examples of correlation lengths and times

We now discuss two simple examples to show the magnitudes of the correlation lengths (times) introduced by the shaping of the wave fields produced by the optical elements of the apparatus. As formulated by Marathay "..the field from a non-coherent (*uncorrelated)* source acquires coherence (*a finite correlation length)* by the very process of propagation." (see [14], p.105.) This is nothing else than what occurs when a random 'white noise' signal acquires a finite correlation time as a result of passing through a low pass filter, something which can easily be observed with an oscilloscope.

For elastic scattering, a beam collimated with slits of width a separated by a

distance L, will have an angular width of $\Delta\theta \sim a/L$ and a spread in momentum perpendicular to the beam of $\Delta k_y \sim k_x \Delta\theta \sim k_x a/L$. This will result in an uncertainty in momentum transfer q of $\Delta q \sim \Delta k_y$.

On the other hand, seen according to quantum mechanics, a beam going through a slit of width a will have an uncertainty in momentum $\Delta k'_y \sim 1/a$ and this will result in a lateral spreading over a distance $\delta \sim \Delta k'_y L/k_x = L/k_x a$ at a distance L in the x direction. We see that the correlation length $\delta \sim 1/\Delta q$ so that both arguments will lead to the same resolution. In order to put the argument on a firmer footing we need to show that a wave acquires a correlation length δ on passing through a slit of width a. δ is the length associated with a Fraunhofer diffraction pattern of a slit. The problem is then to show that this length can arise in the case of Fresnel diffraction (finite distance L). This is the content of the van-Cittert Zernike theorem ([2], [6], [7]) mentioned above. In this way we can describe the scattering process in terms of diffraction at the defining slits and the correlation length of the incident beam. We will be able to describe the scattering in real space instead of in the Fourier transform space defined by $\overrightarrow{q}$.

For a small angle scattering instrument with wavelength of 10 *Angst.* and collimating slits $2cm$ wide separated by $L = 10m$ the correlation width will be about 1600 *Angst., i.e.* much larger than the wavelength but much smaller than the slit width. Correlations over larger distances than this cannot be observed with the assumed instrument.

Applying the same argument to a chopper for massive particles we see that for a chopper opening T and time of flight t_o we will have $\Delta E/E = 2T/t_o = \Delta\omega/\omega$, $\left(\hbar\omega = E = mv^2/2\right)$ by the usual classical arguments. Quantum mechanically, according to the uncertainty principle, passing through a chopper of width T will result in a spread of energy given by $\Delta\omega' \sim 1/T$ and this spread in energy will result in a spread in arrival times $\Delta t/t_o \sim \Delta v/v \sim \Delta\omega'/2\omega \sim 1/2\omega T$. Thus we should expect a correlation time of $\Delta t \sim t_o/2\omega T \sim 1/\Delta\omega$. For a time of flight instrument with pulse width $T = 10^{-5}s$ and a flight path of $L = 5m$, neutron wavelength $\lambda = 5$ *Angst.* we obtain $\Delta t \sim 30ps$ and this is the limit to the correlation times which can be observed with such an instrument.

2 A new look at scattering from many particle systems

We will now show how the effects discussed above are contained in a quantum-mechanical calculation of the scattering. Following the usual treatment of scattering [9], sec. 2.1, [15] the scattered wave is given by

$$\psi_{sc}\left(\overrightarrow{r},t\right) = \int G_o\left(\overrightarrow{r} - \overrightarrow{r_s}, t - t_s\right) V\left(\overrightarrow{r_s},t_s\right) \psi_{sc}\left(\overrightarrow{r_s},t_s\right) d^3 r_s dt_s \tag{1}$$

where $\psi_{sc}\left(\overrightarrow{r_s}\right)$ is the exact solution for the wave function at the position $\overrightarrow{r_s}$, and G_o is the Green's function for the unperturbed problem ($V = 0$). The notation is explained in figs. 4 and 6. As is usual in van Hove scattering we make the Born approximation by replacing $\psi_{sc}\left(\overrightarrow{r_s}\right)$ by the incident wave at the position of the scattering sample, $\psi_{in}\left(\overrightarrow{r_s}\right)$ and take (for the case of neutron scattering)

$$V\left(\overrightarrow{r_s},t\right) = \sum_i \frac{2\pi\hbar^2}{m} b_i \delta\left(\overrightarrow{r_s} - \overrightarrow{r_i}\left(t\right)\right) \equiv \frac{2\pi\hbar^2}{m} \overline{b} \rho\left(\overrightarrow{r_s},t\right) \tag{2}$$

(for coherent scattering), *i.e.* the Fermi pseudo potential where $\overrightarrow{r_i}$ is the position of the i^{th} nucleus, the sum is over all nuclei in the scattering system and $\rho\left(\overrightarrow{r_s},t\right)$ as

defined in (2) is the particle density of the scatterer.[1]

$$\psi_{sc}\left(\overrightarrow{r},t\right)=\int G_o\left(\overrightarrow{r}-\overrightarrow{r_s},t-t_s\right)V\left(\overrightarrow{r_s},t_s\right)\psi_{in}\left(\overrightarrow{r_s},t_s\right)d^3r_sdt_s \tag{3}$$

The integration is carried out over the sample volume and the time interval during which the sample is exposed to the beam.

Equation (3) describes the scattered wave as a superposition of waves produced by many scattering events each occurring at the different space time points $(\overrightarrow{r_s},t_s)$(Fig. 1). This is in contrast to the usual van Hove treatment [1], where one begins with the Fermi golden rule for time dependent perturbations and shows, replacing the energy conservation delta function by its Fourier transform, that the probability of scattering is given by the space-time Fourier transform of the correlation function of $V\left(\overrightarrow{r_s},t_s\right)$. In the appendix to [4] we have shown how one can derive the van Hove result in a more direct way. Introducing $\overrightarrow{r_s}-\overrightarrow{r_s}'=\overrightarrow{\delta}$, $t_s-t_s'=\tau$ we have from (3) for the intensity at the detector, $(\overrightarrow{r_d},t_d)$:

$$\begin{aligned}\left|\psi_{sc}\left(\overrightarrow{r_d},t_d\right)\right|^2&=\int d^3r_sdt_s\int d^3r_s'dt_s'G_o\left(\overrightarrow{r_d}-\overrightarrow{r_s},t_d-t_s\right)G_o^*\left(\overrightarrow{r_d}-\overrightarrow{r_s'},t_d-t_s'\right)\times\\&\times V\left(\overrightarrow{r_s},t_s\right)V^*\left(\overrightarrow{r_s'},t_s'\right)\psi_{in}\left(\overrightarrow{r_s},t_s\right)\psi_{in}^*\left(\overrightarrow{r_s'},t_s'\right)\\&=\int d^3r_sdt_s\int d^3\delta d\tau G_o\left(\overrightarrow{r_d}-\overrightarrow{r_s},t_d-t_s\right)G_o^*\left(\overrightarrow{r_d}-\overrightarrow{r_s}+\overrightarrow{\delta},t_d-t_s+\tau\right)\\&\times V\left(\overrightarrow{r_s},t_s\right)V^*\left(\overrightarrow{r_s}-\overrightarrow{\delta},t_s-\tau\right)\psi_{in}\left(\overrightarrow{r_s},t_s\right)\psi_{in}^*\left(\overrightarrow{r_s}-\overrightarrow{\delta},t_s-\tau\right)\end{aligned} \tag{4}$$

Substituting the Fermi potential we find

$$\left|\psi_{sc}\left(\overrightarrow{r_d},t_d\right)\right|^2=\int d^3r_sdt_s\int d^3\delta d\tau G_o\left(\overrightarrow{r_d}-\overrightarrow{r_s},t_d-t_s\right)G_o^*\left(\overrightarrow{r_d}-\overrightarrow{r_s}+\overrightarrow{\delta},t_d-t_s+\tau\right) \tag{5}$$

$$\times G_s\left(\overrightarrow{\delta},\tau\right)R_{in}\left(\overrightarrow{\delta},\tau\right) \tag{6}$$

Here

$$G_s\left(\overrightarrow{\delta},\tau\right)=\left\langle\rho\left(\overrightarrow{r_s},t_s\right)\rho\left(\overrightarrow{r_s}-\overrightarrow{\delta},t_s-\tau\right)\right\rangle_s \tag{7}$$

is the (van Hove) density-density correlation function and is independent of $\overrightarrow{r_s}$, t_s for the usual case of a homogeneous system. The result depends on the pair correlation function (7) due to the fact that the intensity is proportional to the square of the amplitude as can readily be seen in the above derivation. Because we restrict ourselves to the Born approximation the result is independent of higher order correlation functions.

$$R_{in}\left(\overrightarrow{\delta},\tau\right)=\left\langle\psi_{in}\left(\overrightarrow{r_s},t_s\right)\psi_{in}^*\left(\overrightarrow{r_s}-\overrightarrow{\delta},t_s-\tau\right)\right\rangle_{in} \tag{8}$$

[1] In the interests of simplicity we omit prefactors and constants in the body of this work concentrating on those terms which are essential to the physics. We give a complete treatment in the appendix where we also show how to extract the cross section from our results.

is the auto-correlation function of the incident beam (See fig.2 for the time independent case.). The brackets $\langle\rangle_s$ and $\langle\rangle_{in}$ indicate statistical averages over the sample and incoming beam ensembles respectively.

$R_{in}\left(\overrightarrow{\delta},\tau\right)$ will be seen to be responsible for the contributions of the incident beam to the resolution of the measurement, as, according to (5), the intensity only depends on the correlation function G_s at values of $\left(\overrightarrow{\delta},\tau\right)$ where R_{in} is significant.

The wave function incident on the sample ψ_{in}, is given by

$$\psi_{in}\left(\overrightarrow{r_s},t_s\right)=\int d^2r_o dt_o G_o\left(\overrightarrow{r_s}-\overrightarrow{r_o},t_s-t_o\right)\psi_o\left(\overrightarrow{r_o},t_o\right) \tag{9}$$

where $\psi_o\left(\overrightarrow{r_o},t_o\right)$ is the incident wave function at the entrance of the apparatus[2]. We then find

$$R_{in}\left(\overrightarrow{\delta},\tau\right)=\int d^2r_o dt_o G_o\left(\overrightarrow{r_1},t_s-t_o\right)G_o^*\left(\overrightarrow{r_1}-\overrightarrow{\delta},t_1-\tau\right) \tag{10}$$

where $\overrightarrow{r_1}=\overrightarrow{r_s}-\overrightarrow{r_o}$; $t_1=t_s-t_o$ and we have made the assumption that the incident beam is completely uncorrelated, *i.e.* $\left\langle\psi_o\left(\overrightarrow{r_o},t_o\right)\psi_o^*\left(\overrightarrow{r_o}',t_o'\right)\right\rangle_{in}=\delta^3\left(\overrightarrow{r_o}-\overrightarrow{r_o}'\right)\delta\left(t_o-t_o'\right)$ as is usual in the derivation of the van Cittert-Zernike theorem [2],[6] and [7]. By the Wiener-Khintchine theorem [6] we see this is equivalent to $\left|A\left(\overrightarrow{k}\right)\right|^2=const$, *i.e.* the incident spectrum is broad compared to that selected by the instrument.

In a scattering experiment we measure the integral of $\left|\psi_{sc}\left(\overrightarrow{r_d},t_d\right)\right|^2$ over the entrance area of the detector and, in the case of a time of flight experiment, a definite time interval, dt_d. From (5) the intensity at the detector, I_d, is given by

$$\int d^2r_d dt_d\left|\psi_{sc}\left(\overrightarrow{r_d},t_d\right)\right|^2=\int d^3r_s dt_s\int d^3\delta d\tau R_{out}\left(\overrightarrow{\delta},\tau,\overrightarrow{r_2},t_2\right)G_s\left(\overrightarrow{\delta},\tau\right)R_{in}\left(\overrightarrow{\delta},\tau,\overrightarrow{r_1},t_1\right) \tag{11}$$

where

$$R_{out}\left(\overrightarrow{\delta},\tau,\overrightarrow{r_2},t_2\right)=\int d^2r_d dt_d G_o\left(\overrightarrow{r_2},t_2\right)G_o^*\left(\overrightarrow{r_2}+\overrightarrow{\delta},t_2+\tau\right) \tag{12}$$

and $\overrightarrow{r_2}=\overrightarrow{r_d}-\overrightarrow{r_s}$, $t_2=t_d-t_s$.

Although (11) contains an explicit dependence on only space and time variables, the dependence on the energy and momentum transfer will be seen to emerge from the variation of the phases of the G_o functions.

Now we see there is a symmetry between the integral over the detector parameters, $(\overrightarrow{r_d},t_d)$, of the product of Green's functions depending on $(\overrightarrow{r_2},t_2)$, $R_{out}\left(\overrightarrow{\delta},\tau\right)$, (equ. 12) and the integral over the input slit $(\overrightarrow{r_o},t_o)$ of the Green's function product

[2]Strictly speaking we have $\psi_{in}\left(\overrightarrow{r_s},t_s\right)=\int d^2r_o dt_o\frac{\partial\widetilde{G}_o\left(\overrightarrow{r_s}-\overrightarrow{r_o},t_s-t_o\right)}{\partial\overrightarrow{n}}\psi_o\left(\overrightarrow{r_o},t_o\right)$ where $\widetilde{G}_o$is a Green's function satisfying the boundary condition $\widetilde{G}_o=0$ on the surface of integration, and $\partial/\partial\overrightarrow{n}$ is the gradient in the direction of the surface normal [18],[3]. However this only introduces a change in the prefactor. See the appendix for a complete treatment.

depending on $(\overrightarrow{r_1}, t_1)$, $R_{in}\left(\overrightarrow{\delta}, \tau\right)$, (equ. 10) so that we only have to do the calculation of one of the pairs, say the latter. We then obtain the former integral by applying the symmetry rules:

$$\overrightarrow{\delta} \rightarrow -\overrightarrow{\delta},\ \tau \rightarrow -\tau,\ t_2 \rightarrow t_1,\ \overrightarrow{r_2} \rightarrow \overrightarrow{r_1},\ \overrightarrow{r_s} \rightarrow -\overrightarrow{r_s},\ t_s \rightarrow -t_s. \tag{13}$$

While functions such as $R_{in,out}\left(\overrightarrow{\delta}, \tau\right)$ are usually called 'correlation functions' they really represent a loss of correlation as their arguments increase. In the present context it is instructive to keep this in mind by referring to them as '(loss of) correlation functions'.

Thus while $R_{in}\left(\overrightarrow{\delta}, \tau, \overrightarrow{r_1}, t_1\right)$ represents the loss of correlation between pairs of rays starting at the entrance slit $(\overrightarrow{r}_o)$, propagating to neighboring space time points in the sample $(\overrightarrow{r_s}, t_s)$, $(\overrightarrow{r_s}', t_s')$, $R_{out}\left(\overrightarrow{\delta}, \tau\right)$ represents the loss of correlation due to the difference in path lengths in propagating from two neighboring points in the sample $(\overrightarrow{r_s}, t_s)$, $(\overrightarrow{r_s}', t_s')$ to the detector $(\overrightarrow{r}_d, t_d)$. $G_s\left(\overrightarrow{\delta}, \tau\right)$ is the loss of correlation between the points $(\overrightarrow{r_s}, t_s)$, $(\overrightarrow{r_s}', t_s')$ due to the internal dynamics of the scattering system (See fig. 3). Implicit in the above discussion is the fact that the detector responds to the value of $|\psi_{sc}|^2$ at a single point integrated over the detector area.

The above discussion brings out the fact that the scattering process is intrinsically an interference phenomenon, the cross section being a measure of the loss of correlation between waves following paths separated by $\left(\overrightarrow{\delta}, \tau\right)$ at the sample.

In this paper we limit ourselves to pure coherent scattering. The separation of the scattering into a coherent and an incoherent part can be carried out exactly as in the conventional treatment. [1], [9]. Also in the interests of simplicity we choose to work with a real 'hydrodynamic' density function $\rho\left(\overrightarrow{r}, t\right)$. Replacement of this by $\sum_i \delta\left(\overrightarrow{r} - \overrightarrow{r}_i(t)\right)$ yields results consistent with the conventional approach [1], (see also [4]).

3 Elastic Scattering in Real Space

In this section we specialize to the case of scattering from a static system. We will consider time dependent systems in the following sections.

Using the known form of the Green's function for the time independent Schrödinger equation,

$$G_o = e^{ik|\overrightarrow{r} - \overrightarrow{r_s}|} / |\overrightarrow{r} - \overrightarrow{r_s}|$$

we calculate the input product of Green's functions

$$\sim e^{ik\left(|\overrightarrow{r}_s - \overrightarrow{r_o}| - \left|\overrightarrow{r}'_s - \overrightarrow{r_o}\right|\right)} \tag{14}$$

by expanding

$$|\overrightarrow{r}_s - \overrightarrow{r}_o| - |\overrightarrow{r}'_s - \overrightarrow{r}_o| = \frac{\overrightarrow{\delta} \cdot (\overrightarrow{r}_s - \overrightarrow{r}_o)}{|\overrightarrow{r}_s - \overrightarrow{r}_o|} \approx \frac{\overrightarrow{\delta} \cdot (\overrightarrow{r}_s - \overrightarrow{r}_o)}{|\overrightarrow{r}_s|} \approx \frac{\overrightarrow{\delta} \cdot (\overrightarrow{r}_s - \overrightarrow{r}_o)}{|\overrightarrow{\underline{r}}_1|} \tag{15}$$

which holds to first order in $\overrightarrow{\delta} = \overrightarrow{r}_s - \overrightarrow{r}'_s$. $\overrightarrow{\underline{r}}_1 = \overrightarrow{\underline{r}}_s$ is the location of the center of the sample and we rewrite $\overrightarrow{r}_s \Rightarrow \overrightarrow{\underline{r}}_s + \overrightarrow{\epsilon}_s$ where $\overrightarrow{\epsilon}_s$ is now a small quantity (see fig. 4). Then

$$R_{in}\left(\overrightarrow{\delta}\right) = e^{ik\frac{\overrightarrow{\delta}\cdot(\underline{\overrightarrow{r}}_s+\overrightarrow{\epsilon}_s)}{r_1}}\int d^2r_o e^{-ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{r}_o}{r_1}} = e^{ik\frac{\overrightarrow{\delta}\cdot(\underline{\overrightarrow{r}}_s+\overrightarrow{\epsilon}_s)}{r_1}}\int_{-a}^{a} dy_o e^{-ik\frac{\delta_y y_o}{r_1}} =$$

$$= e^{i\underline{\overrightarrow{k}}_1\cdot\overrightarrow{\delta}} e^{ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{\epsilon}_s}{r_1}} 2a\left(\frac{\sin k\frac{\delta_y a}{r_1}}{k\frac{\delta_y a}{r_1}}\right) \tag{16}$$

where we have taken a one dimensional slit of width $2a$ and $\underline{\overrightarrow{k}}_1 = k\ \underline{\overrightarrow{r}}_1/r_1$ and $r_i = |\underline{\overrightarrow{r}}_i|$. δ_y is the component of $\overrightarrow{\delta}$ perpendicular to the incident beam (see fig.4).

The result (16) is known as the van Cittert-Zernike theorem, [2], [6], [7] and has the form of the Fraunhofer formula. Note that this formula is applicable to geometries where one would not normally observe a Fraunhofer diffraction pattern, except in the case of a spherical wave converging on the point of observation. In applications to optics the phase factors in (16), depending on the position of the pair of points with respect to the center of the sample, as well as on the distance between them are often neglected so that the correlation functions can be considered as real functions depending only on the distance between the two points. However the phase factors play a crucial role in scattering, bringing in the influence of the sample size on the resolution and the dependence on the energy and momentum transfer, as shown below.

By symmetry (13) we can write

$$R_{out}\left(\overrightarrow{\delta}\right) = e^{-i\underline{\overrightarrow{k}}_2\cdot\overrightarrow{\delta}} e^{ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{\epsilon}_s}{r_2}} 2d\left(\frac{\sin k\frac{\delta'_y d}{r_2}}{k\frac{\delta'_y d}{r_2}}\right) \tag{17}$$

where δ'_y represents the component of $\overrightarrow{\delta}$ in the direction perpendicular to the direction of the scattered beam, $\underline{\overrightarrow{k}}_2 = k\underline{\overrightarrow{r}}_2/r_2$ and d is the width of the detector slit (see fig. 4).

Then writing the steady state form of (11) we have

$$I_d = \int d^3\epsilon_s \int d^3\delta R_{out}\left(\overrightarrow{\delta}, \overrightarrow{r_2}\right) G_s\left(\overrightarrow{\delta}\right) R_{in}\left(\overrightarrow{\delta}, \overrightarrow{r_1}\right) \tag{18}$$

Substituting (16) and (17) in (18) we have

$$I_d = \int d^3\epsilon_s \int d^3\delta e^{-i\underline{\overrightarrow{q}}\cdot\overrightarrow{\delta}} e^{ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{\epsilon}_s}{r'}} 2d\left(\frac{\sin k\frac{\delta_{y'} d}{r_2}}{k\frac{\delta_{y'} d}{r_2}}\right) G_s\left(\overrightarrow{\delta}\right) 2a\left(\frac{\sin k\frac{\delta_y a}{r_1}}{k\frac{\delta_y a}{r_1}}\right); \qquad \frac{1}{r'} = \frac{1}{r_1} + \frac{1}{r_2} \tag{19}$$

The term $\exp\left(ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{\epsilon}_s}{r'}\right)$ comes from the phase factor which is normally neglected in optics. (*Note that* $\underline{\overrightarrow{q}} = \underline{\overrightarrow{k}}_2 - \underline{\overrightarrow{k}}_1$, a point which will be discussed in the next section.) In the present case this factor means that moving the location of the correlation regions defined by the two $\sin x/x$ functions (varying $\overrightarrow{\epsilon}_s$) in (19) causes an additional difference in the two optical paths going from the entrance slit to the detector slit via the two points separated by $\overrightarrow{\delta}$, *i.e.* the two paths in fig. 4. This optical path difference represents an additional loss in correlation as the pair of points separated by a constant $\overrightarrow{\delta}$ move away from the optical axis and determines the influence of the sample slit width ($2b$) on the resolution.

3.1 Small angle scattering

For simplicity we will now consider the case of small angle scattering where the only significant component of $\overrightarrow{\delta}$ is $\delta_y = y_s - y'_s$ and $\delta_y \approx \delta_{y'}$.

Performing the ϵ_s integration in (19) over a slit of width $2b$ $\left(\int_{-b}^{b} d\epsilon_s\right)$ defining the sample size yields

$$I_d = \int d^3\delta e^{-i\overrightarrow{q}\cdot\overrightarrow{\delta}} G_s\left(\overrightarrow{\delta}\right) 2d\left[\frac{\sin k\frac{\delta_{y'}d}{r_2}}{k\frac{\delta_{y'}d}{r_2}}\right] 2b\left[\frac{\sin k\frac{\delta_y b}{r'}}{k\frac{\delta_y b}{r'}}\right] 2a\left[\frac{\sin k\frac{\delta_y a}{r_1}}{k\frac{\delta_y a}{r_1}}\right] \tag{20}$$

where we have neglected the extension of the sample in the z direction.

For a, b, d small, corresponding to a high $\overrightarrow{q}$ resolution, the functions in square brackets approach unity for the values of δ for which G_s is significant, and we have the usual result

$$I_d \sim S\left(\overrightarrow{q}\right) = \int d^3\delta e^{-i\overrightarrow{q}\cdot\overrightarrow{\delta}} G_s\left(\overrightarrow{\delta}\right) \tag{21}$$

The same result is obtained if we take the incident and outgoing states as plane waves. The result of an experiment for non-negligible a, b, d must then be found by taking the convolution of (21) with an instrumental resolution function. In the approach of (20) however, the resolution is contained in the (loss of) correlation functions in the square brackets. Equ. (20) clearly shows how correlations between points in the sample separated by larger values of δ contribute less to the intensity. However due to the nature of the $\sin x/x$ functions there are some contributions to the scattering from these larger values, and some regions of δ contribute with a negative sign. These effects, at the 10% level, which do not appear if we approximate the $\sin x/x$ functions by Gaussians of an appropriate width as is often done in practice, become more important for measurements at poor resolution such as in quasi-elastic line broadening.

3.1.1 Comparison with conventional treatment

We see that equ. (20) is the Fourier transform with respect to $\overrightarrow{\delta}$ of a product of $G\left(\overrightarrow{\delta}\right)$ with a function $H\left(a, b, \delta_y\right)$ which is itself the product of three functions,$h_2\left(d, \delta'_y\right)$, $h'\left(b, \delta_y\right)$and $h_1\left(a, \delta_y\right)$. Thus the result (20) is the convolution of the Fourier transform of $G\left(\overrightarrow{\delta}\right)$, *i.e.* $S\left(\overrightarrow{q}\right)$, with the Fourier transform of H. The latter is the convolution of the Fourier transforms of each of the h functions

$$\eta_a\left(q_y\right) = \int d^3\delta e^{-i\overrightarrow{q}\cdot\overrightarrow{\delta}} h_1\left(a, \delta_y\right) = \frac{r_1}{k}\left[u\left(q_y + \frac{ka}{r_1}\right) - u\left(q_y - \frac{ka}{r_1}\right)\right] \tag{22}$$

(r_1/k is a normalizing factor so that $\eta_a\left(q_y\right)$ is normalized to $2a$ and $u\left(x\right)$ is the unit step function). In $\eta_b\left(q_y\right)$ the term $\left(a/r_1\right)$ is replaced by $\left(b/r'\right)$ and in $\eta_d\left(q_y\right)$ it is replaced by $\left(d/r_2\right)$. The convolution of two rectangular functions in q_y, η_a and η_b, yields a trapezoid with a base bounded by

$$\left|q_y\right| < k\left(a/r_1 + b\left(1/r_1 + 1/r_2\right)\right)$$

and a top bounded by

$$\left|q_y\right| < k\left|a/r_1 - b\left(1/r_1 + 1/r_2\right)\right|,$$

just what is expected on geometrical grounds for a collimator consisting of two slits of width $2a$ and $2b$ separated by a distance x as seen by a point r_d away from the second slit (see fig. 5). The subsequent convolution with $\eta_d(q_y)$ yields the effect of finite detector width. From (20) the correlation length of the input function h_1 is seen to be $y_c = r_1/ka$, while (22) gives the width y_q in q, to be $y_q = ka/r_1$. Hence $y_c \cdot y_q = 1$ *or* $y_c \cdot y_p = \hbar$, where the momentum width $y_p = \hbar y_q$.

Thus our result (20) is identical with the usual treatment which yields the convolution of $S(\overrightarrow{q})$ with the resolution function but our view of the scattering is quite different. We see the scattering process as the interaction with the sample of a state whose wave function has a certain spatial auto-correlation function determined by the optical properties of the devices used to define the incoming beam. This has the effect that only pairs of scattering points whose separation is less than the correlation length of the incident beam contribute significantly to the scattered wave and pairs with different separations $\overrightarrow{\delta}$ are weighted by the auto-correlation function of the incoming beam evaluated at $\overrightarrow{\delta}$. Analogous statements apply to the outgoing beam.

Similar considerations have been used by many people (*e.g.* [8]) as a means of deconvoluting the instrument resolution from the measured data. However, in the work cited no mention is made of the physical significance of the Fourier transform of the instrument resolution function (our $H(\delta)$ is called $R(k)$ in ([8])) as the (loss of) correlation functions of the incident and scattered wave functions. In the above treatment we see how the resolution function is the result of the optical properties of the devices defining the incident and scattered beams.

3.1.2 Discussion of the result for small angle scattering

In the previous section we have seen that the contribution to $\left\langle \left|\psi_{sc}(\overrightarrow{r_d})\right|^2 \right\rangle$ from the correlation function of the incident beam represents the effect on the resolution due to the collimation of the incident beam and the correlation function of the scattered beam results in that part of the resolution due to the angular resolution of the scattered beam. Each element of the optical system defining the beam is seen to contribute a factor to the overall correlation function.

In the following we try to illustrate the relation between our treatment of the scattering problem and the conventional approach by concentrating on the contribution of the incident beam to the resolution. If we write

$$\Psi_{inc}(\overrightarrow{r_s}) = \int d^3k_1 A\left(\overrightarrow{k}_1\right) e^{i\overrightarrow{k_1}\cdot\overrightarrow{r_s}} \tag{23}$$

then we have

$$\begin{aligned}\left\langle \Psi_{inc}(\overrightarrow{r_s})\,\Psi^*_{inc}\left(\overrightarrow{r'_s}\right)\right\rangle_b &= \int d^3r_s \int d^3k_1 \int d^3k_1' \left\langle A(k_1)\,A^*\left(k_1'\right)\right\rangle e^{i\overrightarrow{k_1}\cdot\overrightarrow{r_s}-\overrightarrow{k_1'}\cdot\overrightarrow{r_s'}} \\ &= \int d^3k_1 \left\langle \left|A(k_1)\right|^2\right\rangle e^{i\overrightarrow{k_1}\cdot\left(\overrightarrow{r_s}-\overrightarrow{r_s'}\right)}\end{aligned} \tag{24}$$

Substituting this into (18) and writing $\psi_{out} = e^{i\overrightarrow{k}_2\cdot\overrightarrow{r}}$ we obtain

$$\int d^3\epsilon_s \int d^3k_1 \left\langle \left|A(k_1)\right|^2\right\rangle \int d^3\delta e^{-i\overrightarrow{q}\cdot\overrightarrow{\delta}} G_s\left(\overrightarrow{\delta}\right) \sim \int d^3k_1 \left\langle \left|A(k_1)\right|^2\right\rangle S(\overrightarrow{q}) \tag{25}$$

In this case we have taken $\overrightarrow{q} = \overrightarrow{k_2} - \overrightarrow{k_1}$ and we obtain $S(\overrightarrow{q})$ averaged over the momentum spectrum of the incident beam.

In the previous section we took $\overrightarrow{q} = \overrightarrow{\underline{k}}_2 - \overrightarrow{\underline{k}_1}$, *i.e.* the difference between the nominal final wave vector, and the nominal incoming wave vector. By this we mean the wave vector corresponding to a central ray of the incident or scattered beam. Then, in that case, the distribution in $\overrightarrow{k_1}$, or what contains the same information, the correlation function of the incident beam produced by the optical properties of the defining slits, gives the usual result of $S\left(\overrightarrow{q}\right)$ convoluted with an instrumental resolution function. On the other hand, in this section by taking

$$\overrightarrow{q} = \overrightarrow{k_2} - \overrightarrow{k_1} \tag{26}$$

we have assumed perfect knowledge of $\overrightarrow{k_1}$, $\overrightarrow{k_2}$ and we complete the calculation (equ. 25) by integrating over all values of $\overrightarrow{q}$ corresponding to the spread in $\overrightarrow{k_1}$ values in the incident beam. This will of course yield the same results as the previous treatment.

3.2 Elastic scattering at arbitrary angle

We will assume a cylindrical shaped sample (radius R_s) with its axis perpendicular to the scattering plane and thus consider the problem as a two dimensional one. Then the integral over the sample volume in (19) can be written as

$$\begin{aligned}\int d^3\epsilon_s e^{ik\frac{\overrightarrow{\delta}\cdot\overrightarrow{\epsilon}_s}{r'}} &= l\int_0^{R_s}\epsilon_s d\epsilon_s\int_0^{2\pi}d\theta e^{ik\frac{\delta\epsilon_s\cos\theta}{r'}} = l\int_0^{R_s}\epsilon_s d\epsilon_s 2\pi J_0\left(k\frac{\delta\epsilon_s}{r'}\right) = \\ &= 2\pi R_s^2\frac{J_1\left(k\frac{\delta R_s}{r'}\right)}{k\frac{\delta R_s}{r'}}l \end{aligned}\tag{27}$$

where l represents the dimension of the sample perpendicular to the scattering plane. The final result is then

$$\int d^2 r_d\left|\psi_{sc}\left(\overrightarrow{r_d}\right)\right|^2 = \int d^3\delta e^{-i\overrightarrow{q}\cdot\overrightarrow{\delta}}G_s\left(\overrightarrow{\delta}\right)\mathbb{H}\left(\overrightarrow{\delta}\right) \tag{28}$$

where

$$\mathbb{H}\left(\overrightarrow{\delta}\right) = 2d\left(\frac{\sin k\frac{\delta'_y d}{r_2}}{k\frac{\delta'_y d}{r_2}}\right)2\pi R_s^2\left(\frac{J_1\left(k\frac{\delta R_s}{r'}\right)}{k\frac{\delta R_s}{r'}}\right)2a\left(\frac{\sin k\frac{\delta_y a}{r_1}}{k\frac{\delta_y a}{r_1}}\right)l^3 \tag{29}$$

is the Fourier transform of the resolution function. The terms depending on a, d are identical to those discussed above in connection with small angle scattering (20). The Fourier transform of the term containing R_s is given by

$$\int\delta d\delta d\theta e^{iq\delta\cos\theta}\left(\frac{J_1\left(\alpha\delta\right)}{\alpha\delta}\right) = 2\pi\int d\delta J_0\left(q\delta\right)\left(\frac{J_1\left(\alpha\delta\right)}{\alpha}\right) = \tag{30}$$

$$= \begin{cases}\frac{2\pi}{\alpha^2} = \left(\frac{r'}{kR_s}\right)^2 = const & \left\{q < \frac{kR_s}{r'}\right\} \\ 0 & \left\{q > \frac{kR_s}{r'}\right\}\end{cases} \tag{31}$$

3.2.1 Discussion

Just as in the small angle case the result in the general case (28) is seen as the Fourier transform of the product of G_s with a product of functions, each representing the contribution to the resolution of an individual element of the beam handling system.

Corresponding to the $\sin x/x$ functions in one dimension the function $J_1(x)/x$ in two dimensions represents a square shaped resolution in q but in the latter case the resolution is a function of $|\overrightarrow{q}|$ rather than a component of $\overrightarrow{q}$ in the one dimensional cases.

The term involving $(\delta_y a)$ gives the influence of the width of the entrance slit $(2a)$ on the resolution; it depends on r_1 but not on r_2. The term in $(\delta_{y'} d)$ gives the influence of the detector slit width $(2d)$ on the resolution and depends on r_2. The remaining term gives the influence of the sample size and depends on r' because an uncertainty in position at the sample effects the angular resolution in both arms.

The argument of each term can be written as (δ/δ_{ci}) where the δ_{ci} are correlation lengths corresponding to the size of the correlations that can be observed with a given installation. When any of the terms is small, so is the contribution to the total integral, so that the only significant contribution comes from values of $\overrightarrow{\delta}$ for which each term in (29) is significant. Thus the resolution is determined by the overlap of the different correlation volumes and the measurement can be optimized by choosing all regions as approximately equal.

Equ. (28) has been derived for the case of a single incident energy. It is easy to see that if we took an incident spectrum constant in a region $\pm\Delta K$ that the result will be an additional term $\sin x/x$ with $x = \Delta K(\delta_1 - \delta_2)$, where $\delta_{1,2}$ are the components of $\overrightarrow{\delta}$ along the directions of $k_{1,2}$ respectively. A more detailed discussion including the influence of the scattering angle will be presented in a forthcoming paper.

3.3 Neutron spin echo for elastic scattering.

In neutron spin echo for elastic scattering, the relative phase shift between the two spin states is given by $\overrightarrow{q} \cdot \overrightarrow{\delta_{\mathrm{NSE}}}$ with $\overrightarrow{q} = \overrightarrow{k}_2 - \overrightarrow{k}_1$ [4]. Then the average beam polarization will be given by equ. (25) with $\overrightarrow{\delta}$ replaced by $\left(\overrightarrow{\delta} - \overrightarrow{\delta_{\mathrm{NSE}}}\right)$ and to this order the result is independent of the momentum distribution of the incident beam. The dependence of the spin echo signal on the incident beam comes from the fact that $\overrightarrow{\delta_{\mathrm{NSE}}}$ is a function of $\overrightarrow{k_1}$. We now show this in more detail.

In [4] we have described various forms of spin echo spectrometer from a space-time point of view. According to this picture the first arm of the spectrometer causes the beam incident on the scattering sample to be split in space and time, and this splitting results in the scattered beam sampling the scattering system at points separated by this splitting. In this section we show how the current view point yields the same results.

We note that we are considering only scattering with $\overrightarrow{q} = \widehat{i} q_y$. In the case of spin echo the wave function:

$$\Psi_{inc}(\overrightarrow{r}) = \int d^3k_1 A(k_1) e^{i\overrightarrow{k_1}\cdot\overrightarrow{r}} \tag{32}$$

represents the beam arriving at the sample in the absence of the NSE magnetic field. Turning on the first spin echo field (length L) introduces a phase shift which 'splits' the beam on the sample [4]. Calling the components of the split beam on the sample $\psi_2^{\pm}(\overrightarrow{r_s})$ we have:

$$\psi_2^{\pm}\left(\overrightarrow{r_s}\right) = e^{ik_o y_s}\int dk_1 A\left(k_1\right) e^{ik_1 y_s} e^{\pm ik_1\delta_{\rm NSE}/2} e^{\pm i\omega_z L/v_o}$$
$$= \Psi_{\,inc}\left(\overrightarrow{r_s} \pm \delta_{\rm NSE}/2\right) e^{\pm i\omega_z L/v_o} \tag{33}$$

Since the measured quantity in spin echo $\langle\sigma_x\rangle$ depends on the cross correlation between $\psi^{\pm}$ we calculate this cross correlation for the wave functions (33)

$$\left\langle \psi_2^{+}\left(\overrightarrow{r_s}\right)\psi_2^{-*}\left(\overrightarrow{r_s'}\right)\right\rangle = \left\langle \Psi_{\,inc}\left(\overrightarrow{r_s} + \delta_{\rm NSE}/2\right)\Psi^{\,*}_{inc}\left(\overrightarrow{r_s'} - \delta_{\rm NSE}/2\right)\right\rangle$$
$$= R_{in-in}\left(r_s - r_s' + \delta_{\rm NSE}\right) = R_{in-in}\left(\delta + \delta_{\rm NSE}\right) \tag{34}$$

where $R_{in-in}\left(\delta\right)$ is the auto-correlation function of the incident beam in the absence of NSE field.

Now following (3) the wave function of neutrons scattered by the sample that would reach the detector in the absence of a field in the second coil is:

$$\psi_{sc}^{\pm}\left(\overrightarrow{r_d}\right) = \frac{e^{ik_2 r_{\rm d}}}{r_{\rm d}}\int d^3 r_s e^{-i\overrightarrow{k_2}\cdot\overrightarrow{r_s}}\rho\left(\overrightarrow{r_s}\right)\psi_2^{\pm}\left(\overrightarrow{r_s}\right) \tag{35}$$

where $\left|\overrightarrow{k_2}\right| = \left|\overrightarrow{k_1}\right| = k_o$ (elastic scattering) and $\overrightarrow{k_2}$ is directed along a line from the center of the sample to a point on the detector, $\overrightarrow{r_d}$, so that $k_o r_{\rm d} = \overrightarrow{k_2}\cdot\overrightarrow{r_d}$. Thus integrating over d^3k_2 will be equivalent to integrating over the detector area.

When the second NSE coil is turned on, the beam reaching the detector will have an additional phase shift $\left(e^{\mp i\omega_z L/v_o}e^{\mp ik_2\delta_{\rm NSE}/2}\right)$ so that at the detector we will have:

$$\psi_d^{\pm}\left(\overrightarrow{r_d}\right) = e^{i\overrightarrow{k_2}\cdot\overrightarrow{r_d}}\int d^3 r_s e^{-i\overrightarrow{k_2}\cdot\overrightarrow{r_s}} e^{\mp ik_2\delta_{\rm NSE}/2}\rho\left(\overrightarrow{r_s}\right)\Psi_{\,inc}\left(\overrightarrow{r_s} \pm \delta_{\rm NSE}/2\right) \tag{36}$$

Then

$$\langle\sigma_x\rangle = \left\langle\psi_d\left(\overrightarrow{r_d}\right)\left|\sigma_x\right|\psi_d\left(\overrightarrow{r_d}\right)\right\rangle = \int d^3k_2\int d^3 r_s\int d^3 r_s' e^{i\overrightarrow{k_2}\cdot\left(\overrightarrow{r_s'} - \overrightarrow{r_s}\right)} e^{-ik_2\delta_{\rm NSE}}\ldots$$
$$\cdots\rho\left(\overrightarrow{r_s}\right)\Psi_{\,inc}\left(\overrightarrow{r_s} + \delta_{\rm NSE}/2\right)\rho\left(\overrightarrow{r_s'}\right)\Psi^{*}_{\,inc}\left(\overrightarrow{r_s'} - \delta_{\rm NSE}/2\right)$$

The integration over d^3k_2 gives $\delta\left(\overrightarrow{r_s'} - \overrightarrow{r_s} - \delta_{\rm NSE}\right)$ so that we have finally

$$\langle\sigma_x\rangle = \int d^3 r'\left\langle\rho\left(\overrightarrow{r_s}\right)\rho\left(\overrightarrow{r_s} + \delta_{\rm NSE}\right)\right\rangle_s\left\langle\left|\Psi_{\,inc}\left(\overrightarrow{r_s} + \delta_{\rm NSE}/2\right)\right|^2\right\rangle_b \tag{37}$$
$$= G\left(\delta_{\rm NSE}\right)R_{\text{in-in}}\left(0\right) = G\left(\delta_{\rm NSE}\right)$$

Thus we see that in the case considered here with the range of available $\overrightarrow{k_2}$ large enough, and the approximation that $\delta_{\rm NSE}$ is independent of $\overrightarrow{k_{2,1}}$ we get the exact correlation function, independent of the properties of the incident beam. Of course we know this from the classical treatment.

4 Scattering from a time dependent system

In the present section we will show how the ideas presented in the previous section can be applied to scattering from systems fluctuating in time. In scattering from such systems the energy of the scattering radiation is changed (inelastic or quasi-elastic scattering). For simplicity, in this section we will neglect the position dependence of the scattering, considering the scattering system to be concentrated at a single point, $\overrightarrow{r_s}$ and restrict ourselves to massive particle scattering where the energy is defined by choppers.[3] This is equivalent to calculating the scattering integrated over all values of $\overrightarrow{q}$. We will treat the case of time and position dependent scattering in the next section.

Under these restrictions equ. (1) can be written:

$$\psi_{sc}\left(\overrightarrow{r_d},t_d\right)=\int dt_s G_o\left(\overrightarrow{r_d}-\overrightarrow{r_s},t_d-t_s\right)V\left(\overrightarrow{r_s},t_s\right)\psi_{in}\left(\overrightarrow{r_s},t_s\right) \tag{38}$$

for the wave function at the detector located at $\overrightarrow{r_d}$ at time t_d where

$$G_o\left(\overrightarrow{r_d}-\overrightarrow{r_s},t_d-t_s\right)=e^{i\frac{m\left|\overrightarrow{r_d}-\overrightarrow{r_s}\right|^2}{2\hbar\left(t_d-t_s\right)}} \tag{39}$$

is the Green's function (neglecting an unimportant prefactor) for the time dependent unperturbed Schrödinger equation.

The beam will be taken to be incident on the scatterer through a slit (chopper) located at the origin that opens during the time interval $-T\leq t_o\leq T$. (See fig. 6 for a definition of the notation used here.) Analogous to the elastic case above, we consider the wave function incident on the chopper to be completely uncorrelated for different values of t_o. That is, we assume that the correlation time of the beam incident on the chopper is much shorter than the correlation time that will be imposed on the beam by the action of the chopper, or, in other words, the beam incident on the chopper contains a much broader energy spectrum than will be selected by the chopper system.

The wave function leaving the chopper and arriving at a point $\overrightarrow{r_s}$ at time t_s is given by [17], [18], [3]

$$\psi_{in}\left(\overrightarrow{r_s},t_s\right)=\int_{-T}^{T}dt_o e^{i\frac{mr_s^2}{2\hbar\left(t_s-t_o\right)}}\psi_o\left(t_o\right) \tag{40}$$

and

$$\begin{aligned}\left\langle\psi_{in}\left(\overrightarrow{r_s},t_s\right)\psi_{in}^*\left(\overrightarrow{r_s},t_s'\right)\right\rangle&=e^{-i\underline{\omega_1}\tau}e^{i2\underline{\omega_1}\xi_s\frac{\tau}{\underline{t}_1}}\int_{-T}^{T}dt_o e^{-i2\underline{\omega_1}\tau t_o/\underline{t}_1}\\&=e^{-i\underline{\omega_1}\tau}e^{i2\underline{\omega_1}\xi_s\frac{\tau}{\underline{t}_1}}2T\frac{\sin\alpha_1 T}{\alpha_1 T}\end{aligned} \tag{41}$$

where $\underline{\omega_1}=mr_s^2/2\hbar\underline{t}_1^2$

This emergence of plane wave-like states from the time dependent Green's function has been discussed in [16].

[3]The use of crystals as energy defining elements has been briefly discussed according to the present viewpoint in ref. [13] and will be discussed in more detail in a later work.

In deriving (41) we have taken

$$\langle \psi_o(t_o)\, \psi_o^*(t_o')\rangle = \delta(t_o - t_o')$$

This (41) is the time analogue of the van Cittert-Zernike theorem (16) for matter-wave optics.

Following arguments analogous to those in the previous section we find for the intensity at the detector integrated over a time channel of width $2T_d$:

$$\int_{-T_d}^{T_d} dt_d\, |\psi_{sc}(\overrightarrow{r_d}, t_d)|^2 = \int_{-T_s}^{T_s} d\xi_s \int d\tau e^{i\omega\tau} e^{i\mu\xi_s} G_s(\tau)\, 2T \frac{\sin\alpha_1 T}{\alpha_1 T} 2T_d \frac{\sin\alpha_2 T_d}{\alpha_2 T_d} \quad (42)$$

where

$$\alpha_1 = \frac{2\underline{\omega_1}\tau}{\underline{t_1}}, \quad \alpha_2 = \frac{2\underline{\omega_2}\tau}{\underline{t_2}}, \quad \mu = \alpha_1 + \alpha_2 \quad (43)$$

and $\omega = \underline{\omega_2} - \underline{\omega_1}$ is the mean energy transfer of the scattered particles. There is a second chopper placed close to the scatterer which opens for $\underline{t_s} - T_s \le t_s \le \underline{t_s} + T_s$. The integral over $d\xi_s$ is seen to give $2T_s \frac{\sin\mu T_s}{\mu T_s}$ with

$$\mu = 2\tau \left(\frac{\underline{\omega_2}}{\underline{t_2}} + \frac{\underline{\omega_1}}{\underline{t_1}} \right) \quad (44)$$

The term $e^{i\mu\xi_s}$ comes from the phase factor which is normally neglected in optics. In the present case this factor has the effect that moving the correlation interval (τ) (varying ξ_s) in (42) causes an additional difference in the two optical paths going from the entrance chopper to the detector chopper via the two points separated by τ, *i.e.* the paths *1* and *2* in fig. 6). This defines the influence of the sample chopper width ($2T_s$) on the resolution.

We then have:

$$\int_{-T_d}^{T_d} dt_d\, |\psi_{sc}(\overrightarrow{r_d}, t_d)|^2 = 2T_s 2T 2T_d \int d\tau e^{i\omega\tau} G_s(\tau)\, \mathbb{H} \quad (45)$$

$$\mathbb{H} = \frac{\sin\alpha_2 T_d}{\alpha_2 T_d} \frac{\sin\mu T_s}{\mu T_s} \frac{\sin\alpha_1 T}{\alpha_1 T} \quad (46)$$

where we have written $G_s(\tau)$ for $\langle \rho(\overrightarrow{r_s}, t_s)\, \rho^*(\overrightarrow{r_s}, t_s - \tau)\rangle_s$ the time dependent density-density correlation function of the scattering system.

Thus the scattering cross section is found to be proportional to the Fourier transform of the density-density correlation function of the scattering system multiplied by a function of τ which is the product of three functions representing the effects of the opening times of the three choppers on the resolution of the measurement. In this case the scattering is seen as the interaction of the incoming state with the time fluctuations of the scattering system. The wave function of the incoming state has an auto-correlation function determined by the (time-dependent) optical properties of the defining choppers. Only pairs of scattering events separated by times τ, less than the correlation time of the incoming wave contribute to the scattered wave and their contribution is weighted by the beam auto-correlation function evaluated at τ.

4.1 Discussion

The situation represented by (45) is completely analogous to the elastic scattering case (28). The opening times $T_{1,2}$ influence the resolution by an amount depending on $\underline{t}_{1,2}$ respectively, while T_s contributes to the uncertainty in the travel times in both arms and depends on a weighted combination of the two travel times. The argument of each term can be written as τ/τ_{ci} where the τ_{ci} are different correlation times. Clearly the measurement will be optimized when all the τ_{ci} are approximately equal.

5 A space and time dependent system

Rewriting equ. (11) we have for the intensity at the detector:

$$\int d^2 r_d dt_d \left|\psi_{sc}\left(\overrightarrow{r_d}, t_d\right)\right|^2 = \int d^3 r_s dt_s \int d^3\delta d\tau R_{out}\left(\overrightarrow{\delta}, \tau, \overrightarrow{r_2}, t_2\right) G_s\left(\overrightarrow{\delta}, \tau\right) R_{in}\left(\overrightarrow{\delta}, \tau, \overrightarrow{r_1}, t_1\right) \tag{47}$$

with G_o given by equ. 39. We recall that

$$\overrightarrow{r_s'} = \overrightarrow{r_s} - \overrightarrow{\delta}, \quad t_s' = t_s - \tau \tag{48}$$

and we expand

$$\left|\overrightarrow{r_s'} - \overrightarrow{r_o}\right|^2 \approx \left|\overrightarrow{r_s} - \overrightarrow{r_o}\right|^2 - 2\overrightarrow{\delta}\cdot\left(\overrightarrow{r_s} - \overrightarrow{r_o}\right) \tag{49}$$

$$\frac{1}{t_s' - t_o} \approx \frac{1}{t_s - t_o} + \frac{\tau}{\left(t_s - t_o\right)^2} \tag{50}$$

where we have neglected terms of the order τ^2 and δ^2. To calculate the product of the two Green's functions in (10) we need to calculate the quantity

$$\Lambda_{in} = \frac{\left|\overrightarrow{r_s} - \overrightarrow{r_o}\right|^2}{\left(t_s - t_o\right)} - \frac{\left|\overrightarrow{r_s'} - \overrightarrow{r_o}\right|^2}{\left(t_s' - t_o\right)} = \tag{51}$$

$$= \frac{\left|\overrightarrow{r_s} - \overrightarrow{r_o}\right|^2}{\left(t_s - t_o\right)} - \left(\left|\overrightarrow{r_s} - \overrightarrow{r_o}\right|^2 - 2\overrightarrow{\delta}\cdot\left(\overrightarrow{r_s} - \overrightarrow{r_o}\right)\right) \times \left[\frac{1}{t_s - t_o} + \frac{\tau}{\left(t_s - t_o\right)^2}\right] \tag{52}$$

$$= -\frac{\left|\overrightarrow{r_s} - \overrightarrow{r_o}\right|^2 \tau}{\left(t_s - t_o\right)^2} + \frac{2\overrightarrow{\delta}\cdot\left(\overrightarrow{r_s} - \overrightarrow{r_o}\right)}{\left(t_s - t_o\right)} \tag{53}$$

Using our usual notation

$$t_s \Rightarrow \underline{t}_s + \xi_s = \underline{t}_1 + \xi_s \tag{54}$$

$$\overrightarrow{r_s} \Rightarrow \overrightarrow{\underline{r}_s} + \overrightarrow{\epsilon_s} = \overrightarrow{\underline{r}}_1 + \overrightarrow{\epsilon_s} \tag{55}$$

we have

$$\Lambda_{in} = -\frac{|\overrightarrow{r_s} - \overrightarrow{r_o}|^2 \tau}{\underline{t}_1^2}\left(1 - 2\frac{\xi_s - t_o}{\underline{t}_1}\right) + \frac{2\overrightarrow{\delta}\cdot(\overrightarrow{r_s} - \overrightarrow{r_o})}{\underline{t}_1}\left(1 - \frac{\xi_s - t_o}{\underline{t}_1}\right) \tag{56}$$

and finally

$$\Lambda_{in} = -\frac{|\overrightarrow{\underline{r}}_1|^2 \tau}{\underline{t}_1^2} + \frac{2\overrightarrow{\delta}\cdot(\overrightarrow{\underline{r}}_1)}{\underline{t}_1} + t_o\left(-\frac{2|\overrightarrow{\underline{r}}_1|^2\tau}{\underline{t}_1^3} + \frac{2\overrightarrow{\delta}\cdot\overrightarrow{\underline{r}}_1}{\underline{t}_1^2}\right) + \overrightarrow{r_o}\cdot\left(\frac{2\overrightarrow{\underline{r}}_1\tau}{\underline{t}_1^2} - \frac{2\overrightarrow{\delta}}{\underline{t}_1}\right) \tag{57}$$

$$+\xi_s\left(-\frac{2\overrightarrow{\delta}\cdot(\overrightarrow{\underline{r}}_1)}{\underline{t}_1^2} + \frac{2|\overrightarrow{\underline{r}}_1|^2\tau}{\underline{t}_1^3}\right) + \overrightarrow{\epsilon_s}\cdot\left(\frac{2\overrightarrow{\delta}}{\underline{t}_1} - \frac{2\overrightarrow{\underline{r}}_1\tau}{\underline{t}_1^2}\right) \tag{58}$$

Thus the phase of the integrand of R_{in} (equ. 10) is:

$$\frac{m}{2\hbar}\Lambda_{in} = -\underline{\omega}_1\tau + \overrightarrow{\underline{k}}_1\cdot\overrightarrow{\delta} + (t_o - \xi_s)\,\alpha_1 + (\overrightarrow{r_o} - \overrightarrow{\epsilon_s})\cdot\overrightarrow{\beta_1} \tag{59}$$

where

$$\underline{\omega}_1 = \frac{m}{2\hbar}\frac{|\overrightarrow{\underline{r}}_1|^2}{\underline{t}_1^2}, \quad \overrightarrow{\underline{k}}_1 = \frac{m}{\hbar}\frac{\overrightarrow{\underline{r}}_1}{\underline{t}_1} \tag{60}$$

$$\alpha_1 = \left(-\frac{2\underline{\omega}_1\tau}{\underline{t}_1} + \frac{\overrightarrow{\delta}\cdot\overrightarrow{\underline{k}}_1}{(\underline{t}_1)}\right), \quad \overrightarrow{\beta_1} = \left(\frac{\overrightarrow{\underline{k}}_1\tau}{\underline{t}_1} - \frac{m\overrightarrow{\delta}}{\hbar(\underline{t}_1)}\right) \tag{61}$$

Then

$$R_{in} = \int_{y=-a}^{y=a} d^2r_o \int_{-T}^{T} dt_o e^{i\frac{m}{2\hbar}\Lambda_{in}} = e^{i\left(-\underline{\omega}_1\tau + \overrightarrow{\underline{k}}_1\cdot\overrightarrow{\delta}\right)} e^{-i\left(\xi_s\alpha_1 + \overrightarrow{\epsilon_s}\cdot\overrightarrow{\beta}_1\right)} 2al\frac{\sin\beta_{1y}a}{\beta_{1y}a} 2T\frac{\sin\alpha_1 T}{\alpha_1 T} \tag{62}$$

where we have specialized to the case of a one dimensional input slit going from $y = -a$ to $y = a$. $l(\gg a)$ is the height of the slit. Note that $\overrightarrow{\underline{k}}_1$ is defined to be parallel to the x axis so that $\left(\overrightarrow{\underline{k}}_1\right)_y = 0$. This result (62) is the generalization of the van Cittert-Zernike theorem to the case of time dependent matter wave optics. Comparison with the pure spatial case (16) and the pure time dependent case (41) shows that these are special limits of the general case (62).

By our symmetry rules (13) R_{out} can be written

$$\int_{Det} d^2r_d \int_{-T_d}^{T_d} dt_d e^{i\frac{m}{2\hbar}\Lambda_{out}} = e^{i\left(\underline{\omega}_2\tau - \overrightarrow{\underline{k}}_2\cdot\overrightarrow{\delta}\right)} e^{i\left(\xi_s\alpha_2 - \overrightarrow{\epsilon_s}\cdot\overrightarrow{\beta}_2\right)} 2dl\frac{\sin\beta_{2y'}d}{\beta_{2y'}d} 2T_d\frac{\sin\alpha_2 T_d}{\alpha_2 T_d} \tag{63}$$

where y' indicates the direction perpendicular to the output arm ($\overrightarrow{\underline{r}}_2 = \overrightarrow{\underline{r}}_d - \overrightarrow{\underline{r}}_s$) and

$$\alpha_2 = \left(\frac{2\underline{\omega}_2 \tau}{\underline{t}_2} - \frac{\overrightarrow{\delta} \cdot \overrightarrow{\underline{k}}_2}{\underline{t}_2} \right), \quad \overrightarrow{\beta_2} = \left(-\frac{\overrightarrow{\underline{k}}_2 \tau}{\underline{t}_2} + \frac{m\overrightarrow{\delta}}{\hbar \underline{t}_2} \right) \tag{64}$$

Performing the integral in (47) over the sample volume and scattering time yields

$$\int d^3\epsilon_s \int_{-T_s}^{T_s} d\xi_s \cdots = 2T_s \frac{\sin \mu T_s}{\mu T_s} l \int_0^{R_s} \epsilon_s d\epsilon_s \int_0^{2\pi} d\theta e^{i\epsilon_s \nu \cos\theta} \tag{65}$$

where we assumed the sample to be in the shape of a cylinder of radius R_s. In equ. (65)

$$\mu = \alpha_2 - \alpha_1 = \left(-\overrightarrow{\delta} \cdot \left(\frac{\overrightarrow{\underline{k}}_1}{\underline{t}_1} + \frac{\overrightarrow{\underline{k}}_2}{\underline{t}_2} \right) + 2\tau \left(\frac{\underline{\omega}_1}{\underline{t}_1} + \frac{\underline{\omega}_2}{\underline{t}_2} \right) \right) \tag{66}$$

and

$$\nu = \left| \overrightarrow{\beta}_2 - \overrightarrow{\beta}_1 \right| = \left| \frac{m\overrightarrow{\delta}}{\hbar} \left(\frac{1}{\underline{t}_1} + \frac{1}{\underline{t}_2} \right) - \tau \left(\frac{\overrightarrow{\underline{k}}_1}{\underline{t}_1} + \frac{\overrightarrow{\underline{k}}_2}{\underline{t}_2} \right) \right| \tag{67}$$

The two dimensional integral gives

$$2\pi \int_0^{R_s} \epsilon_s d\epsilon_s J_0(\nu\epsilon_s) = 2\pi R_s^2 \frac{J_1(\nu R_s)}{\nu R_s} \tag{68}$$

The result of evaluating (47) is then

$$\int d^3 r_d dt_d \left| \psi_{sc}(\overrightarrow{r_d}, t_d) \right|^2 = \int d^3\delta d\tau e^{i\left(\omega\tau - \overrightarrow{q}\cdot\overrightarrow{\delta}\right)} G_s\left(\overrightarrow{\delta}, \tau\right) \mathbb{H} \tag{69}$$

where

$$\omega = \underline{\omega}_2 - \underline{\omega}_1, \quad \overrightarrow{q} = \overrightarrow{\underline{k}}_2 - \overrightarrow{\underline{k}}_1 \tag{70}$$

$$\mathbb{H} = 2^5 \left(d\frac{\sin\beta_{2y'}d}{\beta_{2y'}d} \right) \left(T_d \frac{\sin\alpha_2 T_d}{\alpha_2 T_d} \right) \left(T_s \frac{\sin\mu T_s}{\mu T_s} \right) \left(2\pi R_s^2 \frac{J_1(\nu R_s)}{\nu R_s} \right) \left(T_o \frac{\sin\alpha_1 T_o}{\alpha_1 T_o} \right) \left(a\frac{\sin\beta_{1y}a}{\beta_{1y}a} \right) \tag{71}$$

5.1 Discussion

For convenience we rewrite below

$$\alpha_1 = \frac{\overrightarrow{\underline{k}}_1}{\underline{t}_1} \cdot \left(\overrightarrow{\delta} - \overrightarrow{v}_1 \tau \right), \quad \overrightarrow{\beta_1} = -\frac{m}{\hbar \underline{t}_1} \left(\overrightarrow{\delta} - \overrightarrow{v}_1 \tau \right)$$

$$\alpha_2 = -\frac{\overrightarrow{\underline{k}}_2}{\underline{t}_2} \cdot \left(\overrightarrow{\delta} - \overrightarrow{v}_2 \tau \right), \quad \overrightarrow{\beta_2} = \frac{m}{\hbar \underline{t}_2} \left(\overrightarrow{\delta} - \overrightarrow{v}_2 \tau \right) \tag{72}$$

$$\mu = (\alpha_2 - \alpha_1), \quad \nu = \left| \overrightarrow{\beta}_2 - \overrightarrow{\beta}_1 \right|$$

α_1 and β_1 represent the influence of the input slit and chopper on the overall resolution, μ and ν the influence of the sample size and chopper while α_2 and β_2 represent the effects of the detector slit and time resolution.

Each of these variables has a term proportional to τ and a term proportional to δ. Keeping in mind the Fourier transform (69) we see that the terms proportional to τ will give the energy resolution and those proportional to δ will give the q resolution. Thus, for example, the τ term in α_1 represents the effects of the first chopper opening time, T_o, on the energy resolution just as in the pure time dependent case, equations (43, 45) while the δ term represents the effect of the first chopper width on the q resolution, *i.e.* the chopper width results in an uncertainty in $\overrightarrow{k}_1$ that gives a contribution to the q resolution. Similar remarks hold for each of the terms in (72). Note that by definition $\overrightarrow{\underline{k_1}}_y, \overrightarrow{\underline{k_2}}_{y'} = 0$ but we display these terms in (72) to keep the symmetry.

We see that our approach yields directly the mutual influence of the q and ω resolutions in a compact analytic form. Since each element of the apparatus is associated with a definite factor in $\mathbb{H}$, we see directly the influence of the individual elements on the overall resolution. Modification of these elements can readily be accommodated as shown in the next section.

If we wish we can follow the more usual procedure of replacing each term by a Gaussian function with the appropriate width. This then allows an easy way to estimate the overall width of the ω, q resolution (given by the convolution of the Fourier transform of each term in $\mathbb{H}$) as the square root of the sum of the squares of the widths of each term and shows that the optimum is when all the widths are equal.

Each factor in (71) is associated with a certain velocity in $\left(\overrightarrow{\delta}, \tau \right)$ space so that the relevant correlation volumes can be considered as moving as a function of τ. For example the volume associated with α_2 is moving with a velocity $\overrightarrow{\underline{v}}_2$, the nominal velocity of the scattered wave. Thus we see that the coherence volume associated with the detector parameters can be considered as consisting of the region limited by $\pm \hbar \underline{t}_2 / md$ in the δ'_y direction and $\pm \underline{t}_2 / T_d k_2$ in the $\delta_{x'}$ direction, moving (as function of τ) with velocity v_2 in the x' direction (direction of the scattered beam). Each of the other pairs of terms contributes a similar moving limit to the region of $\left(\overrightarrow{\delta}, \tau \right)$ space.

In a forthcoming work we will present a detailed discussion of the shapes and behavior of these correlation volumes for a series of instruments.

5.2 Entrance slit with moving edges

We now modify the above treatment to consider the more realistic case where the chopper slits move with finite speed. See [3] for discussion of a related problem. We consider that the slit width depends on the time t_o, at which the beam passes through the slit in the following manner:

$$a\left(t_o\right)=\begin{cases}0 & if\, t_o<-T_1\\ a+\theta t_o\, if\, -T_1\le t_o\le 0\\ a-\theta t_o\, if\, 0<t_o\le T_1\\ 0 & if\, T_1<t_o\end{cases} \tag{73}$$

where a is the maximum opening of the slit (which occurs at $t_o=0$) and $\theta=a/T_1$. Then we rewrite equ. (62) as

$$R_{in}=e^{i\Gamma}l\int_{-T_o}^{T_o}dt_o e^{i\alpha t_o}\int_{-a(t_o)}^{a(t_o)}dy e^{i\beta_{1y}y} \tag{74}$$

$$=e^{i\Gamma}l\int_{-T_o}^{T_o}dt_o e^{i\alpha t_o}\frac{\left[e^{i\beta_{1y}a(t_o)}-e^{-i\beta_{1y}a(t_o)}\right]}{i\beta_{1y}} \tag{75}$$

where Γ represents those terms in equ. (57) which do not depend on t_o and r_o. The last integral can be evaluated to give

$$R_{in}=e^{i\Gamma}l\left[2aT_o\frac{\sin\frac{1}{2}\left(\alpha_1T_o-\beta_{1y}a\right)}{\frac{1}{2}\left(\alpha_1T_o-\beta_{1y}a\right)}\frac{\sin\frac{1}{2}\left(\alpha_1T_o+\beta_{1y}a\right)}{\frac{1}{2}\left(\alpha_1T_o+\beta_{1y}a\right)}\right] \tag{76}$$

The rest of the calculation proceeds as above so the only change in the result is to replace the $\sin x/x$ functions depending on a, T_o and their associated prefactors in (71) with the function in square brackets in (76). The correlation function (76) is seen to have the correct normalization at $\alpha_1=\beta_{1y}=0$, *i.e.* at $\overrightarrow{\delta}=\tau=0$. The two terms have separated maxima occurring at

$$\delta_x\pm\delta_y\frac{a}{\underline{v}_1T_o}-\underline{v}_1\tau=0 \tag{77}$$

where δ_x is the component of $\overrightarrow{\delta}$ along the direction of the incident beam and δ_y its component in the perpendicular direction. Thus for $\tau=0$ the maxima lie on the lines through the origin:

$$\frac{\delta_y}{\delta_x}=\pm\frac{\underline{v}_1T_o}{a}=\pm\frac{\underline{v}_1}{v_s} \tag{78}$$

where v_s is the velocity of the slit boundary. For non-zero τ the slope of the lines stays the same but the crossing point moves along the δ_x axis with a velocity $\underline{v}_1$. The correlation volume for the input beam, *i.e.* the overlap region where the two factors in (76) are significant, is thus seen to be given by a complex interplay between the slopes of the lines and the widths of the $\sin x/x$ functions which depend on the correlation lengths $\delta_{yc}=L_1/\underline{k}_1a,\ \delta_{xc}=\underline{t}_1/T_o\underline{k}_1$, and the correlation time $\tau=\underline{t}_1/2\underline{\omega}_1T_o$.

This is a demonstration of some of the advantages of the present method, showing how a change of a component can be easily accommodated and how complex relationships between the energy and momentum resolution can be represented in a straight forward manner.

6 Conclusions

Starting with Fermi's 'golden rule' for perturbation theory and assuming the incident and final wave functions of the scattered particles to be plane waves, van Hove [1] showed that the scattering cross section is proportional to a function of the momentum and energy transfer, $S(q,\omega)$, which in turn is the Fourier transform of the time and space dependent auto-correlation function of the density fluctuations of the scattering system.

However since no experiment is ever done with perfect resolution, measurements yield the convolution of $S(q,\omega)$ with an instrument resolution function. The determination of the resolution and its use in the interpretation of the data are left to the experimenters.

In this conventional approach a scattering experiment is divided up into two separate parts which are treated by two, different, mutually exclusive approximations. The scattering event in the sample is treated by assuming infinitely extended plane waves for the incident and scattered beams, while the beam is treated as consisting of classical point particles in order to describe its motion through the apparatus. This same, dual approach is also used to discuss multiple scattering: the scattering events are described as a scattering of plane wave states while the motion between two scattering points is treated as a classical trajectory. We hope to present a unified treatment of multiple scattering using the present approach in the near future.

In the present work, the entire scattering experiment is described as a wave-optical phenomenon. This more general approach provides a rigorous justification for the usual dual approximation since, in most cases the correlation volumes are much smaller than the geometrical dimensions (beam cross sections, chopper pulse lengths, etc.) involved. However in some cases this is not true and the complete wave-optical description of the global system of instrument and scattering sample must be used. In addition to precision light scattering experiments such cases include perfect crystal neutron optics (*e.g.* interferometry) where the definition of the wave vector is so precise that the correlation lengths can be comparable to the geometrical dimensions. These cases are sometimes referred to as "spherical wave effects"[19]. In fact these effects are natural consequences of the full wave-optical analysis presented here.

Usual methods of determining the resolution include measurement of a known calibration sample, Monte Carlo calculations and estimates based on assuming a Gaussian form for the separate contributions to the overall resolution so that the separate contributions can be combined using the sum of squares rule.

One method of carrying out the deconvolution is to divide the Fourier transform of the measured data by the Fourier transform of the resolution function. Usually however a model form is assumed for $S(\overrightarrow{q},\omega)$ and some parameters of the model are varied until the convolution with the resolution function gives agreement with the measured data.

In the present work we calculate the incoming wave function as it is shaped by transmission through the elements of the apparatus according to the laws of optics. For example on propagating through a slit and/or a chopper a wave acquires a correlation function with a finite width. After scattering, the individual waves scattered from a neighboring pair of points in the sample lose correlation on passing through the post-sample elements of the apparatus. We have shown that the probability of a scattered particle arriving at a detector at a given space-time location is proportional to the Fourier transform of the density fluctuation auto-correlation function (as shown by van Hove) but multiplied by a function $\mathbb{H}\left(\overrightarrow{\delta},\tau\right)$ which represents the

resolution of the measurement and is the Fourier transform of the instrument resolution as it is normally considered. This function is the product of a series of functions, each representing the effect of a single element of the apparatus on the measurement. Changes in one element can be easily accommodated by changes in the appropriate function. The regions of $\left(\overrightarrow{\delta}, \tau\right)$ space where each function is significant are the correlation volumes for the different elements of the apparatus. A measurement can be optimized by maximizing the region of overlap of all the correlation volumes. The fact that these volumes are seen to be moving as a function of the correlation time τ, is a reflection of the fact that the ω, and $\overrightarrow{q}$ resolutions depend on each other to some extent and we have an analytic description of this dependence. The present analysis offers a graphical method of optimizing each measurement, with the influence of all parameters such as the scattering angle included. In a forthcoming work we will give detailed examples of how this can be carried out for various types of scattering measurements.

7 Appendix - Normalization and calculation of cross sections

In the main text we have neglected all prefactors and normalizing constants in the Green's functions and incident wave function. In this appendix we will show how inclusion of these factors leads to the usual expression for the cross section and give a proper calculation of ψ_{in} as mentioned in footnote 2 of section 2. We will treat the most general case of space and time dependent density fluctuations (sec. 5).

7.1 Normalization of the Green's functions

7.1.1 Calculation of the incident wave.

To calculate the wave function incident on the scattering sample we use

$$\psi_{in}\left(\overrightarrow{x}, t\right) = \frac{-1}{4\pi} \int_0^{t_+} dt_o \int d^2 S_o \psi_o\left(\overrightarrow{x}_o, t_o\right) \overrightarrow{n} \cdot \overrightarrow{\nabla}_o \widetilde{G}\left(\overrightarrow{x} - \overrightarrow{x}_o, t - t_o\right) \tag{79}$$

where the integral over $d^2 S_o$ is taken over the plane of the entrance slit, $z_o = 0$, and ψ_o is non-zero only in the entrance slit ($-a < y_o < a$). (We make the Kirchhoff approximation as is usual in optics). The function $\widetilde{G}$ satisfies the boundary condition $\widetilde{G}\left(z_o = 0\right) = 0$ and can be taken as

$$\widetilde{G} = g\left(\overrightarrow{r}_+, t\right) - g\left(\overrightarrow{r}_-, t\right) \tag{80}$$

where

$$\left(\overrightarrow{r}_\pm\right)^2 = \left(x - x_o^2\right) + \left(y - y_o\right)^2 + \left(z \mp z_o\right)^2 \tag{81}$$

and $g(\overrightarrow{r}, t)$ satisfies

$$\nabla^2 g - \frac{2m}{i\hbar} \frac{\partial g}{\partial t} = -4\pi \delta^{(3)}\left(\overrightarrow{r}\right) \delta(t) \tag{82}$$

$g(\overrightarrow{r}, t)$ is then normalized as [18]:

$$g(\overrightarrow{r}, t) = \left(\frac{2\pi i\hbar}{m}\right) \left(\frac{m}{2\pi i\hbar t}\right)^{3/2} e^{i\frac{mr^2}{2\hbar t}} \tag{83}$$

Then (equ. 79)

$$\psi_{in}\left(\overrightarrow{x},t\right)=z\left(\frac{m}{2\pi i\hbar}\right)^{3/2}\int_{0}^{t+}\frac{dt_o}{\left(t-t_o\right)^{5/2}}\int d^2S_o\psi_o\left(\overrightarrow{x}_o,t_o\right)\underline{G}\left(\overrightarrow{x}-\overrightarrow{x}_o,t-t_o\right) \tag{84}$$

where $\underline{G}=\exp\left(i\frac{mr^2}{2\hbar t}\right)$is the Green's function without prefactor as used in the main text.

7.1.2 Calculation of the scattered wave.

Taking the interaction potential as

$$V\left(\overrightarrow{r},t\right)=2\pi\frac{\hbar^2b}{m}\rho\left(\overrightarrow{r},t\right) \tag{85}$$

where b is the scattering length and ρ is the number density of scatterers the scattered wave is found to be (making the Born approximation)

$$\psi_{sc}\left(\overrightarrow{r},t\right)=\frac{2\pi\hbar b}{im}\int G_o\left(\overrightarrow{r}-\overrightarrow{r}_s,t-t_s\right)\rho\left(\overrightarrow{r}_s,t_s\right)\Psi_{in}\left(\overrightarrow{r}_s,t_s\right)d^3r_sdt_s \tag{86}$$

where G_o satisfies

$$i\hbar\frac{\partial G_o}{\partial t}+\frac{\hbar^2}{2m}\nabla^2G_o=\frac{-\hbar}{i}\delta^{(3)}\left(\overrightarrow{r}\right)\delta(t) \tag{87}$$

and is normalized as [16]

$$G_o=\left(\frac{m}{2\pi i\hbar t}\right)^{3/2}\underline{G} \tag{88}$$

so that (86)

$$\psi_{sc}\left(\overrightarrow{r},t\right)=\left(\frac{2\pi\hbar b}{im}\right)\left(\frac{m}{2\pi i\hbar}\right)^3\frac{z_1}{\underline{t}_2^{3/2}\underline{t}_1^{5/2}}\int\int dt_od^2S_o\int\int\underline{G}\rho\underline{G}\psi_od^3r_sdt_s \tag{89}$$

where $\underline{t}_{1,2}$ are the average flight times in the two arms of the apparatus and z_1 is the distance from the entrance slit to the center of the sample. We have neglected the variation of the prefactors over the regions of integration as these are small compared to the distances and flight times involved. With suitable normalization of the incoming wave, $\psi_{o,}$ the intensity, I_d, counted in the detector during the time interval $2T_d$, will be given by

$$v_2\int d^2r_d\int_{-T_d}^{T_d}dt_d\left|\psi_{sc}\right|^2=v_2b^2\left(\frac{m}{2\pi\hbar}\right)^4\Sigma_o\frac{z_1^2}{\underline{t}_2^3\underline{t}_1^5}l^2\int\int d^3r_sdt_s\int\int d^3\delta d\tau R_{out}\left\langle\rho\rho'\right\rangle R_{in} \tag{90}$$

where $R_{out,in}$ are defined as in section 2 and $\langle \rho\rho' \rangle$ is related to the van Hove correlation function $G_s\left(\overrightarrow{\delta},\tau\right)$ by [1]

$$\langle \rho\rho' \rangle = \rho_s G_s\left(\overrightarrow{\delta},\tau\right) \tag{91}$$

and the correlation function of the input beam is defined to be

$$\langle \psi_o\left(\overrightarrow{r},t\right)\psi_o^*\left(\overrightarrow{r}',t'\right)\rangle_{inc} = \Sigma_o \delta^{(2)}\left(\overrightarrow{r}-\overrightarrow{r}'\right)\delta\left(t-t'\right) \tag{92}$$

l is the height of the beam perpendicular to the scattering plane.

The multiple integral in (90) has been evaluated in (69). Setting all the slit widths and chopper opening times to zero in the function $\mathbb{H}$ so that this takes on its maximum value (note: $\lim_{x\to 0} J_1(x)/x = 1/2$) we get from (90) for the intensity I_d

$$I_d = v_2 b^2 \left(\frac{m}{2\pi\hbar}\right)^4 \Sigma_o \frac{z_1^2}{\underline{t}_2^3 \underline{t}_1^5} l^3 \mathbb{K}\left(\pi R_s^2\right)\rho_s \frac{2\pi}{N} S\left(\overrightarrow{q},\omega\right) \tag{93}$$

where we have used van Hove's definition [1]:

$$S\left(\overrightarrow{q},\omega\right) = \frac{N}{2\pi}\int\int d^3\delta d\tau e^{i\left(\overrightarrow{q}\cdot\overrightarrow{\delta}-\omega\tau\right)} G_s\left(\overrightarrow{\delta},\tau\right) \tag{94}$$

$N = \rho_s\left(\pi R_s^2 l\right)$ is the total number of scattering centers in the sample and

$$\mathbb{K} = \left(2T_o\right)\left(2a\right)\left(2T_s\right)\left(2T_d\right)\left(2d\right) \tag{95}$$

Since $z_1 = v_1\underline{t}_1\ (= L_1)$ we finally obtain:

$$I_d = v_2 b^2 \left(\frac{m}{\hbar}\right)^4 \frac{\Sigma_o}{\left(2\pi\right)^3}\frac{v_1^2}{\underline{t}_2^3\underline{t}_1^3} l^2 \mathbb{K} S\left(\overrightarrow{q},\omega\right) \tag{96}$$

7.2 Calculation of the cross section

7.2.1 Normalization of the input wave correlation function

Rewriting the definition (92) we have

$$\begin{aligned}\langle \psi_o \psi_o^{*\prime}\rangle_{inc} &= \Sigma_o \delta^{(2)}\left(\overrightarrow{r}-\overrightarrow{r}'\right)\delta\left(t-t'\right) = \\ &= \Sigma_o \frac{1}{\left(2\pi\right)^2}\int d^2k e^{i\overrightarrow{k}\cdot\left(\overrightarrow{r}-\overrightarrow{r}'\right)}\frac{1}{2\pi}\int d\omega e^{-i\omega\left(t-t'\right)}\end{aligned} \tag{97}$$

so that

$$\langle\rho\rangle = \left\langle |\psi|^2\right\rangle = \langle \psi_o\psi_o^{*\prime}\rangle_{\delta=\tau=0} = \frac{\Sigma_o}{\left(2\pi\right)^3}\int\int d^2k d\omega \to \infty \tag{98}$$

for the white spectrum we are assuming. From this we conclude that the density fluctuations contained in the interval $d^2k d\omega$ are given by

$$\frac{d^3\rho}{d\omega dk^2}d^2kd\omega = \frac{\Sigma_o}{(2\pi)^3}d^2kd\omega \tag{99}$$

or

$$\frac{d^3\rho}{d\omega dk^2} = \frac{\Sigma_o}{(2\pi)^3} \tag{100}$$

7.2.2 Calculation of the intensity in terms of the cross section.

The incident flux in a velocity interval dv_1 can be written

$$v_1\left(\frac{d^3\rho}{d\omega dk^2}\right)\left(\frac{d\omega}{dv_1}\right)dv_1k_1^2d\Omega_1 \tag{101}$$

with v_1 the incident velocity and $d\Omega_1$ the input solid angle. Multiplying by the chopper opening time $(2T_o)$ and the area of the input slit $(2al)$ we obtain the total number of particles contained in one pulse. The fraction of particles scattered into the velocity range dv_2 and solid angle $d\Omega_2$ is then given by

$$\left(\frac{1}{A_s}\right)\frac{d^2\sigma}{d\omega d\Omega}\frac{d\omega}{dv_2}dv_2d\Omega_2 \tag{102}$$

where in the first factor A_s is the area of the sample exposed to the beam. We use the cross section definition of van Hove which applies to the whole sample (*i.e.* N times the cross section for the individual scattering atoms). Now with

$$\frac{d\omega}{dv} = \frac{mv}{\hbar}, \quad dv_{1,2} = \frac{v_{1,2}2T_{s,d}}{\underline{t}_{1,2}}, \quad d\Omega_1 = \frac{A_s}{L_1^2}, \quad d\Omega_2 = \frac{2dl}{L_2^2} \tag{103}$$

we have

$$v_1\frac{\Sigma_o}{(2\pi)^3}\frac{mv_1}{\hbar}\frac{v_1 2T_s}{\underline{t}_1}k_1^2\frac{A_s}{L_1^2}\left(2T_o\right)\left(2al\right)\left(\frac{1}{A_s}\right)\frac{d^2\sigma}{d\omega d\Omega}\frac{mv_2}{\hbar}\frac{v_2 2T_d}{\underline{t}_2}\frac{2dl}{L_2^2} \tag{104}$$

for the number of scattered particles reaching the detector.
Collecting terms we have

$$\frac{\Sigma_o}{(2\pi)^3}\left(\frac{m}{\hbar}\right)^4\frac{v_1^3}{\underline{t}_1^3\underline{t}_2^3}l^2\mathbb{K}\frac{d^2\sigma}{d\omega d\Omega} \tag{105}$$

Comparing with (96) we see that the two expressions agree if

$$\frac{d^2\sigma}{d\omega d\Omega} = b^2\frac{v_2}{v_1}S\left(\overrightarrow{q},\omega\right) \tag{106}$$

which is equ. (26) of ref. [1].

Figure Captions

Fig.1) Scattering in first Born approximation (equ. 3). The incoming wave is scattered at $\overrightarrow{r}_s$ and the scattered wave propagates according to G_o. The Green's function (39) has the property that at a fixed time the wavelength decreases with distance from the source.

Fig. 2 a) The incident wave field, initially uncorrelated, emerges from the entrance slit with correlations over a length scale d, inversely proportional to the slit width, a.

2 b) Showing the phase relations responsible for the correlations of the wave field according to the van Cittert-Zernike theorem. Waves emerging from the uncorrelated point sources a, b, c arrive at the scattering points r_s, r'_s with different relative phases. The phase difference of the waves arriving at the different scattering points from each source point depends on the location of the source point as well as the distance $\overrightarrow{\delta} = \overrightarrow{r}_s - \overrightarrow{r'}_s$, increasing with $\overrightarrow{\delta}$. This increasing phase difference leads to a decrease in correlation between the waves arriving at the points r_s, r'_s. Because the phase differences between the uncorrelated source points are random they have no influence on the correlation function.

Fig. 3) Illustrating equ. (11). The functions R_{in}, R_{out} represent the effects of the phase differences between the paths 1 and 2 on the correlation functions of the beam. G_s is the correlation of the density fluctuations at the different scattering points and $|\psi_d|^2$ is the sum of the contributions from paths going through all possible pairs of points in the sample. Only pairs whose separation lies within the correlation volumes contribute.

Fig. 4) Elastic scattering - general case. Showing the meaning of the symbols used in the text. $\overrightarrow{\delta}$ is the distance between the two scattering events at $\overrightarrow{r}_s$, $\overrightarrow{r}'_s$. $\overrightarrow{\epsilon}_s$, $\overrightarrow{\epsilon}'_s$ are the positions relative to center of the sample. Further symbols are defined in the text.

Fig. 5) Resolution function for small angle scattering

Fig. 6) Scattering from a time dependent system. The scatterer is assumed to be concentrated at a single point ($\overrightarrow{r}_s$). The resolution is determined by the phase difference between paths (1) and (2). Note the analogy with elastic scattering - fig. 4. τ is the delay between the two scattering events at t_s, t'_s. ξ_s, ξ'_s are the delays relative to the nominal scattering time $\underline{t}_s$.

References

[1] L. van Hove, Phys. Rev., **95**, 249 (1954)

[2] M. Born and E. Wolf, *Principles of Optics*, 6^{th} ed., Pergamon Press, Oxford Sec. 10-4,
L. Mandel and E. Wolf, *Optical coherence and quantum optics,* Cambridge University Press, (1995)

[3] R. Gähler, R. Golub, Z. Phys. **B56**, 5 (1984),
R. Golub, J. Felber, G. Müller and R. Gähler, Physica **B162**, 191 (1990)

[4] R. Gähler, R. Golub, K. Habicht, T. Keller and J. Felber, Physica **B229,1** (1996)

[5] F. Mezei, ed., *Neutron Spin Echo*, Proceedings of a Laue-Langevin Institut Workshop, Oct. 1979. Springer-Verlag, 1980.

[6] D. C. Champeney, *Fourier Transforms and their Physical Applications*, Academic Press, N.Y., 1973

[7] P.H. van Cittert, Physica **1**, 201 (1934)
F. Zernike, Physica **5**, 785 (1938)
H.H. Hopkins, Proc. Roy. Soc. **A208**, 263 (1951)

[8] D. S. Sivia, R. N. Silver and R. Pynn, Nucl. Inst. Meth. **A287**, 538 (1990)

[9] V. F. Sears, *Neutron Optics*, Oxford, (1989), sec. 7.4

[10] R. Golub and S. K. Lamoreaux, Phys. Letts. **A162**, 122 (1992)

[11] F. Mezei in J. P. Hansen, D.Levesque and J. Zinn-Justin, *eds.* Les Houches Session LI, July, 1989; *Liquids, Freezing and Glass Transition*, Elsevier, (1991)

[12] A.Q.R. Baron, A.I. Chumakov, H.F. Grünsteudel, H. Grünsteudel, L. Niesen and R. Rüffer, Phys. Rev. Letts **77**, 4308, (1996)

[13] T. Keller, W. Besenböck, J. Felber, R. Gähler, R. Golub, P. Hank and M. Köppe, *Proceedings of the Intern Conf on Neutron Scattering*, Interlaken (1996), Physica **B234**, 1120 (1997).
J. Felber, R. Gähler, R. Golub, K. Prechtel, "Coherence volumes and neutron scattering", *to be published.*

[14] A.S. Marathay, *Elements of Optical Coherence Theory,* John Wiley, 1982

[15] B. A. Lippman and J. Schwinger, Phys. Rev. **79**, 469 (1950)

[16] R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals*, McGraw-Hill , N.Y. (1965)

[17] M. Moshinsky, Phys. Rev. **88**,625 (1952)

[18] P. M. Morse and H. Feshbach, *Methods of Theoretical Physics*, Mcgraw-Hill, N.Y. (1953), Vol. 1, Chapter 7

[19] C. G. Shull and J. A. Oberteuffer, Phys. Rev. Letts. **29**, 871 (1972)
J. Arthur and M. A. Horne, Phys. Rev. **B32**, 5747 (1985)
J. Arthur, C. G. Shull and A. Zeilinger, Phys. Rev. **B32**, 5753 (1985)

This figure "fig1.gif" is available in "gif" format from:

http://arXiv.org/ps/physics/9712029v1

This figure "fig2.gif" is available in "gif" format from:

http://arXiv.org/ps/physics/9712029v1

This figure "fig3.gif" is available in "gif" format from:

http://arXiv.org/ps/physics/9712029v1

This figure "fig4.gif" is available in "gif" format from:

http://arXiv.org/ps/physics/9712029v1

This figure "fig5.gif" is available in "gif" format from:

http://arXiv.org/ps/physics/9712029v1


This figure "fig6.gif" is available in "gif" format from:
http://arXiv.org/ps/physics/9712029v1